



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Electrodeposition of U and Pu on Thin C and Ti Substrates

R. A. Henderson, J. M. Gostic

May 21, 2010

ILWOG-42

Los Alamos, NM, United States

May 24, 2010 through May 27, 2010

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Electrodeposition of U and Pu on Thin C and Ti Substrates

Roger Henderson & Julie Gostic

LLNL

Abstract

Physics experiments aimed at deducing key parameters for use in a variety of programs critical to the mission of the National Laboratories require actinide targets placed onto various substrates. The target material quantity and the substrate desired depend upon the type of experiment being designed. The physicist(s) responsible for the experimental campaign will consult with the radiochemistry staff as to the feasibility of producing a desired target / substrate combination. In this report we discuss the production of U and Pu targets on very thin C and Ti substrates. The techniques used, plating cells designed for, tips, and limits will be discussed.

Experimental

Prior to electroplating U or Pu isotopes on any substrate, the material is purified. For U, this comprises running two anion columns using Bio-Rad AG-1-X8, 100-140 mesh resin. The first column is loaded from 8M HNO₃, and more 8M HNO₃ is used to run U off of the resin. The second anion column is loaded from 9M HCl, rinsed with more 9M HCl, and the U finally eluted with 0.1M HCl. The U is taken to dryness, then dissolved in a minimum volume of 0.05M HNO₃. This solution is assayed to determine what aliquot is needed to produce the desired target layer.

The Pu chemistry begins with a complete redox cycle to fix the oxidation state of the material into +4. The procedure is to evaporate the source material. The material is then taken to dryness from HCl/HI, followed by HCl/HNO₃. The cycle is repeated, then the source material is precipitated from HCl as the OH⁻. The resulting precipitate is dissolved in a minimum amount of 9M HCl with a couple of drops of HNO₃, and loaded onto AG-1-X8, 100-140 mesh, pre-treated with 9M HCl. The column is rinsed with 9M HCl, and then the Pu is eluted with 1:12 con HI / 10M HCl. The Pu solution is taken to dryness, and then brought up in a minimum amount of 8M HNO₃ plus a drop of saturated NH₄NO₂. A second anion column is loaded from 8M HNO₃, and then rinsed with more 8M HNO₃. The resin is converted to the Cl form with 9M HCl, and Pu is eluted with 1:12 con HI / 10M HCl. The Pu is taken to dryness, and then dissolved in a minimum amount of 0.05M HNO₃. The resulting solution is assayed to determine the aliquot size needed for the desired target layer.

An experimental campaign to determine the (n,f) cross sections for a variety of U isotopes has been undertaken, and employs the surrogate ratio technique which permits the use of more stable isotopes to be utilized for the experiments using (α,α') reactions to produce the same compound nucleus that

would result from the absorption of a neutron on a more radioactive isotope. These experiments are fielded at the LIBERACE instrument at the Lawrence Berkeley Laboratories' 88" cyclotron. The desired



Figure 1 – Exploded drawing view of an electroplating cell designed for use with thin C substrates mounted on Al support frames.

substrates for these experiments were either 60 or 100 $\mu\text{g}/\text{cm}^2$ C foils, which are quite fragile. A specially designed electroplating assembly was created for this project which permitted the target material to be plated onto the C foil already mounted in its' Al support frame, see Figure 1. The base piece and clamp plate are Al; the chimney and support plate are Teflon. The O-ring is rubber. The base plate has two alignment rods which aid in positioning the chimney onto the target frame. The Teflon support plate has a milled slot in it corresponding to the outline of the target support frame.

A target frame with a C foil glued on is carefully slid into place, with the foil on the bottom of the frame, touching the Teflon support plate. This contains the forces laterally across the foil, and minimizes forces perpendicular to the foil, which can easily result in foil rupture. Once in place, the chimney is carefully lowered into position, and the clamp plate attached gently. The screws are driven in with fingers touching only the bit of a screwdriver, not the handle, to better control a very gentle pressure being applied. Isopropyl alcohol is added until the chimney is about $\frac{1}{2}$ full, about 3 mL, and the assembly is checked for leaks. As long as there are no leaks, an aliquot of material to be plated, < 100 μL in 0.05 M HNO_3 , is added to the cell.

The solution in the cell was carefully stirred using a plastic transfer pipette. A 300 W plating power supply, operated in the constant current mode, was set to 4 mA. A Pt anode wire with a small coil at the end is carefully placed into the cell and suspended just above the substrate. The leads to the power supply are hooked to the anode wire and the Al target frame. Plating is allowed to occur for one hour. At the end of the plating process, the power supply is turned off and the electrical leads removed. The solution is carefully removed from the cell with a transfer pipette, being careful not to touch the C substrate. When the majority of the liquid has been removed, the cell is carefully disassembled. The target is then slid back off of the support plate while wet, and placed directly into a plastic 50 mL centrifuge tube. Unlike metal backed targets, these are not baked in an oven or on a hot plate due to the fragility of the substrate material.

Shown in Figure 2 is one example of a plating cell used for depositing material on both sides of a metal substrate simultaneously. The large blocks in the figure are all milled from Teflon. The outer two foils, the cathode materials, are Pt foils. The center target foil is held in place by O-rings on either side of the foil. Alignment

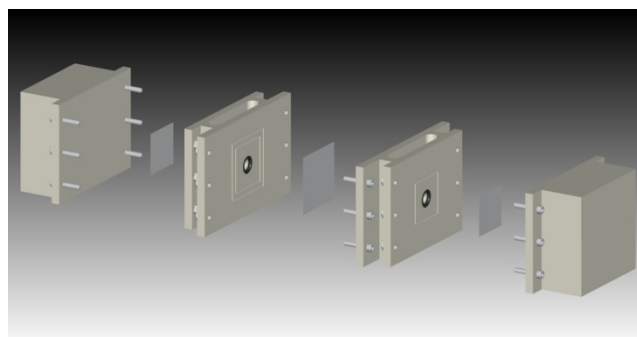


Figure 2 – Exploded drawing of a cell for electroplating both sides of a substrate material simultaneously.

grooves milled into the faces towards the target substrate allow the target to be accurately positioned into the apparatus should multiple layers be required to achieve the desired target thickness. The Pt anode foils are permanently clamped into place, and small Pt disks are used to achieve electrical contact between the foils and the plating power supply.

The cell is assembled carefully and the mounting screws are tightened firmly. About 40 mL of IPA is added to each side of the cell, along with 100 μL of 0.05 M HNO_3 (containing the isotope of interest). A peristaltic pump is used to circulate the solutions in each side of the cell separately. The solution is circulated for about 30 minutes, then pumped out using the peristaltic pump. This process helps to clean the foil prior to electroplating. About 40 mL of fresh IPA is then added to each side of the cell, along with the aliquot of target material to be plated, < 100 μL in 0.05M HNO_3 . The peristaltic pump is started, and the solution is allowed to mix for about 30 minutes prior to the plating supply being turned on. The plating supply is set for a 36 mA current limit for a 4 cm diameter plating area. The anodes have a jumper between them. The anodes and cathodes are connected to the power supply. Plating takes place for 1 hour. The power supply is switched off, and the peristaltic pump is used to remove the solution from the cell. The cell center is disassembled, and the foil carefully removed and placed into a clean glass petri dish. The foil is then heated on a hot plate, cooled, and gamma assayed to determine the thickness of the deposit.

Discussion

It was found during the course of these experiments that the aliquot size, in particular for the small-volume apparatus used for the thin C foil backed targets, had to be less than 100 mL of 0.05M HNO_3 . If the volume of acid used exceeded this value, a very poor, lumpy, deposit resulted. It is not known whether the effect was due in part to the adhesive used to glue the target foil to the frame, but that could be a contributing factor. In any case, ensuring that less than 100 mL of 0.05M HNO_3 was used ensured that the deposit was uniform and smooth.

The plating supply being used in these experiments is a 300 W supply, capable of sustaining a maximum current of 1 A at a maximum voltage of 300 V. Setting the system in constant current mode and limiting the current to 3 mA/cm² of target surface area gives very good results. No attempt was made to determine what an optimal setting might be, this setting worked fine.

U was deposited using both apparatus. In the case of the double-sided targets, the client wished a final target thickness of 400 $\mu\text{g}/\text{cm}^2$ on each side of the foil. An aliquot sufficient to supply this in a single layer was utilized, however after gamma assay of the target produced, only 300 mg/cm² had been plated, indicating a 75% plating efficiency. This figure has been reproducible across the targets produced so far. A second layer was electroplated onto the first, using the same procedure and an aliquot of source material needed to bring the total to 400 mg/cm². This second layer was deposited near quantitatively.

Pu was deposited only on the thin C substrates, and from previous experience with creation of a large ^{239}Pu source, the electroplating can be achieved quantitatively in a single layer of 125 mg/cm^2 , which was the desired aliquot. 4

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.